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# Adsorption of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>(152+154)</sup> Eu onto polyaniline stannic silicomolybdate in aqueous chloride medium

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#### **KEYWORDS ABSTRACT**

Adsorption and Separation of 85Sr, 152+154Eu and activation product of 137Cs from radioactive waste (radioactive waste our Laboratories from Egyption (ET-RR1) research reactor) was achieved, these elements are considered as hazardous materials adsorbed using polyaniline stannic silicomolybdate was developed. Interactions of <sup>137</sup>Cs(I), <sup>152,154</sup>Eu(III), and <sup>85</sup>Sr(II) ions from HCl acid solutions with polyaniline stannic silicomolybdate matrix, dried at 50±0.1°C, have been individually investigated by the batch equilibration method. The sorption behavior of the three ions showed a selectivity sequence in the following order: Cs(I) > Eu(III) > Sr(II). The break through capacities of polyaniline stannic silicomolybdate for <sup>137</sup>Cs(I), <sup>152,154</sup>Eu(III), and 85Sr(II) was determined and the adsorption yield of these isotopes were found to be 94.6%, 92.6% and 87% respectively. The capacity was determined to be 2.5 meq/g from results obtained in Freundlich and D-R isotherms. Adsorption capacities and mean energies calculated from D-R isotherm parameters decreased by increasing ionic strength of the prepared exchanger. The activated energy calculated from Arhenious was found to be 20.7, 18.6,16.9 KJmol<sup>-1</sup> for <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu respectively. © 2015 Trade Science Inc. - INDIA

Adsorption; Polyaniline stannic silicomolybdate; Freundlich and D-R isotherms.

#### INTRODUCTION

Radioactive waste is a source of water pollution. Radioactive substances are coming from fission products where some of them used in, industrial, medical and other scientific processes. There are also naturally occurring radioisotopes from organisms and the environment. The radioactive waste can cause serious environmental pollution. The separation of some isotopes present in radioactive waste must be done to avoid the harmful effect on the human body. Some of these isotopes come from fis-

sion products and neutron activation such as <sup>137</sup>Cs, 85Sr and 152+154Eu. which comes from fission products and radioactive isotopes from our research Laboratories and from Egyption (ET-RR1) research reactor. Different techniques were used for separation processes such as solvent extraction, ion exchange and membranes. Adsorption on Polyaniline stannic silicomolybdate, an a new organic-inorganic composite material was synthesized by mixing polyaniline, an electrically conducting organic polymer into the matrices of inorganic precipitate of stannic silicomolybdate. The experimental parameters

such as mixing volume ratio and pH were established for the synthesis of the material. The material was found granular, thus suitable for column operations. Adsorption of cesium from aqueous solution using cerium molybdate—pan composite In this study, a cerium molybdate-polyacrylonitrile (CM-PAN)(1-12) composite ion exchanger was synthesized. The adsorption of Cs(I) from aqueous solutions by CM-PAN composite was investigated under batch and continuous conditions. The distribution coefficient of cesium on the composite sorbent was studied as a function of pH, solution temperature.the presence of interfering cations and the optimum conditions for a batch system were determined. Pseudo-first- and second-order sorption kinetic models were used to investigate the kinetics of adsorption and the results pointed to the pseudosecond-order model for cesium sorption kinetics(13-18). The intraparticle diffusion model was used to predict rate-limiting step of the ion exchange process in order to specify the sorption mechanism. Langmuir, Freundlich and Dubinin-Radushkevich (D-R)(19) isotherm models were fitted to the experimental sorption data, where the Freundlich model showed a good agreement. The adsorption thermodynamic parameters, standard enthalpy, entropy and Gibbs' free energy were calculated and the reaction was found to be endothermic and spontaneous. Finally, the dynamic sorption capacities of the sorbent at two break throughs were calculated from the continuous system.in the paper described. The goal of this work is adsorption of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>(152+154)</sup> Eu on Stannic Silicomolybdate polymeric adsorbent in aqueous chloride medium, ion exchanger for separation of fission products<sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu has been achieved from chloride medium in various concentrations. Adsorption capacities and mean energies calculated from D-R isotherm parameters decreased by increasing ionic strength on the prepared exchanger.

#### **EXPERIMENTAL**

#### Chemicals and reagents

All the Chemicals and reagents were of analytical grade (AR), they are all B.D.H.

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#### **Preparation of reagents**

Solutions of 0.1 M stannic chloride pentahydrate sodium silicate and sodium molybdate were prepared in demineralized water. Solution by 10% v/v of double distilled aniline and 0.1 M potassium persulphate were prepared in 0.1 M HCl, 0.1 M solution of <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>152</sup>Eu, <sup>60</sup>Co, <sup>103</sup>Ru and <sup>241</sup>Am radioactive waste were prepared in double distilled water.

#### Preparation polyaniline stannic silicomolybdate

Polyaniline gels were prepared by mixing different volume ratio of 10% aniline and 0.1 M potassium persulphste with continuous stirring by magnetic stirrer. Green colored polyaniline gel was obtained by keeping the solution below 10°C for an hour. A precipitate of stannic silicomolybdate was prepared at 25° C by adding mixture of 0.1 M sodium silicate and 0.1 M sodium molybdate solution gradually to an aqueous solution of 0.1 M stannic chloride pentahydrate in different volume ratios(20). The white precipitates were obtained when the pH of the mixture was adjusted to 1.2, 1.4 and 1.5 on addition of HCl with continuous stirring. The gels of polyaniline were then added to the white inorganic precipitate of stannic (IV) silicomolybdate and mixed thourghly under constant stirring. The resultant green colour gel obtained were rept for 24hr at room temperature. The supernatant liquid was filtered off by suction the gel was washed with distilled water till the filtrated became neutral dried in an oven at 50°C. the dried material was broken into small granules on immersion in distilled water and the granules were converted into H<sup>+</sup> from by placing in 0.1 M HNO3 solution for 24 hr. polyaniline stannic silicomolybdate particles size 50-100µm were obtained by sieving.

#### **Characterization of PASSM**

Polyaniline can be easily synthesized chemically from acidic aqueous solutions. In this study, polyaniline gel was prepared by oxidative coupling using  $K_2$   $S_2$   $O_8$  in an acidic aqueous medium at below 10UC as given below(21). Figure 1: Structure formula polyaniline and stannic silicomolybdate(PASSM). The formation of inorganic

Figure 1: Structure formula polyaniline and stannic silicomolybdate(PASSM)

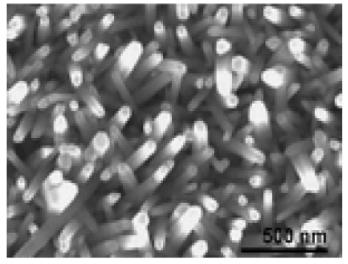


Figure 2: Scanning electron microscope at two different magnifications (500nm and 1000nm)

precipitate stannic silicomolybdate was significantly affected by the pH 1.03 of the mixture, and the most favorable pH of the mixture was 1.4. The preparation of the inorganic precipitate at pH lower or higher than 1.4 lead to decrease in yield and in ion-exchange capacity of the material

#### Scanning Electron Microscopy (SEM)

Scanning electron micrographs of polyaniline stannic silico molybdate. Figure 2: were recorded using scanning electron microscope at two different magnifications (500µm and 1000µm).

#### **Radioactive tracers**

All the tracers used in this thesis were prepared by neutron irradiation of high purity grade target materials. In this respect, suitable weight (20-100) micro gramme of each target was wrapped separately in high purity grade aluminum foil and placed in a separate irradiation cane. These cans were irradiated by neutrons for 48 hours in the ET-RR-1 Egyptian Reactor at Inshas(22). using neutron flux of 1.43x10<sup>13</sup> n.sec<sup>-1</sup>. The radioactive isotopes used in the present study were <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu.

All these isotopes were obtained from Amersham (England). All the pH values measured during the course of this work were accurately determined by employing a pH-meter of type B-417 HANA instrument. For equilibrium experiments a good shaker of the type SANKYO, a Centrifuge of the type UNI-VERSAL with. a muffle furnace from LINDBERG and a special micropipette with disposable tips were used. Tri-Carb 1600 CA liquid scintillation analyzer was used for the determination of  $\beta$ -emission via liquid scintillation solution (Hionic fluxar). Amodel 800A multichannel analyzer consisting of NaI(Ti) activated crystal flat type of 256 channels connected to an automatic scaler was used for counting the gamma activity.

#### Determination of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu

In this concern, 100 ml 0.25 M from NaCl in presence of 0.1 M NaNO<sub>3</sub> at pH~ 6.53 were equilbrirated with 1g of the anionic exchanger stannic silicomolybdate. After the respective equilibration time 2h, 20 ml of the same solution containing the ions under investigations <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu at pH~5.86 was shacked with the applied resin

for two hours, after equilibration, filtration was carried out and the filtration analysis. Showed that <sup>137</sup>Cs absent where the other 2 ions. <sup>152+154</sup>Eu. Were present in the filtrate. This result confirmed the concept that <sup>137</sup>Cs is taken by the resin. In order to obtaining Cs-137 from the used resin (polyaniline stannic silicomolybdate) the loaded resin was shaken with a solution of 0.03 N EDTA acid in presence of 0.1 N NaNO<sub>3</sub> at pH~ 6.53 and studied distribution coefficient has low value reached into =2.21.

#### **Distribution coefficients**

The K<sub>d</sub> values for elements were determined by batch operation. 250mg of ion exchanger were equilibrated for three hours at 25±1°C in 20ml of 2x10<sup>-5</sup>M solution containing the respective radionuclide tracer and the activities of solution after and before were measured by Gamma spectrometer and Alpha counter.

# Adsorption of metals onto polyaniline stannic silicomolybdate

The metal adsorption experiments were designed to provide a quantitative physico-chemical characterization of metal binding by polyaniline stannic silicomolybdate species as a function of pH, the(pH-dependent adsorption edge) and as a function of aqueous metal concentration (adsorption isotherm). In the pH-edge experiments, the initial metal concentration was set at  $(2x10^{-5} - 2x10^{-2}M)$  for Cs>Sr and Eu respectively, while the pH ranged from 1.26 to 6.3 depending on each metal., at constant pH (D-R and Freundlich adsorption isotherm), the metal ion concentration ranged as follows:  $(2x10^{-4} - 2x10^{-3}M)$  for Cs>Sr and Eu.

#### Dubinin-Radushkevich (DR) analysis

Dubinin-Radushkevich (DR) analysis has been devoted to elucidating the micropore structures of various solids regardless of insufficient understanding of the basis of the D-R equation..

#### **Analytical procedure**

For loading radioactive loading, 800ml of the simulated radioactive waste solution containing and studied elements in sodium nitrate 0.25 M, at pH ~1.26 was run down the column. An inductively

coubled plasma-optical emission spectrometry(ICP-OES) of ARL type model 1520, was used for element analysis where argon is used and the power was 1.5 watt and the temperature of the plasma was about 6000°C. All elements were found in the effluent except <sup>137</sup>Cs.

#### **Liquid-solid investigations**

For batch investigations take twenty samples from the mother solution, put in each of 20 ml were adjusted to different pH·s (1.53-10.6). These samples were shaken for two hours with one grams of the exchange. Calculated the distribution coefficient

$$\mathbf{K}_{d} = \frac{\mathbf{C}_{d} - \mathbf{C}}{\mathbf{C}_{x}} \frac{\mathbf{V}}{\mathbf{m}} \tag{1}$$

#### **Column application**

A chromatographic double jacket column was used in connection with a Julabo circulator which control the temperature with an a quracy of  $\pm\,0.01^{\circ}$ C. The column was of 1 cm diameter and 25 cm long and filled with 7.5 gm of either stannic silicomolybdate as anionic exchangers to give ~22 cm bed length. Before use, the column was preconditioned by 300ml of the loading solution and in all cases the follow rate of the mobile phase was 2 ml/min.

#### **Effect of eluent concentration**

To find out the optimum concentration of the eluent for complete elution of H<sup>+</sup> ions, a fixed volume (250 mL) of NaNO<sub>3</sub> solution of different concentrations was passed through the columns, containing 1.0 g of the exchanger in H<sup>+</sup> form with a flow rate of 0.5mL/min<sup>"1</sup>. The effluents were titrated against a standard solution of 0.1M NaOH to find the H<sup>+</sup> ions eluted out.

#### Ion exchange capacity

The ion exchange capacity, which is generally taken as a measure of the hydrogen ion liberated by neutral salt to flow through the composite cation exchanger was determined by standard column process. 1.0 g (dry mass) of the composite ion exchange material in H<sup>+</sup> form was placed in a glass column with a glass wool support at the bottom. It was washed with demineralized water to remove any

excess of acid remained sticking on the particles. The hydrogen ions eluted(22-25) with 0.1M solution of different alkali and alkaline earth salts. The flow rate was kept 0.5 mLmin<sup>"1</sup>. The collected effluent was titrated against a standard solution of sodium hydroxide using phenolphthalein as an indicator. The hydrogen ions released were then calculated. A 1.0 g (dry mass) of stannic Silicomolybdate in the H<sup>+</sup> form was packed in a glass column having a glass wool support at the base. A 0.1 M NaNO3 solution was passed slowly by adjusting the effluent rate at 9–10 drops/min. The effluent was carefully collected in a 250 ml conical flask. The complete replacement of H<sup>+</sup> from the ion exchanger by <sup>137</sup>Cs was checked by comparing the pH of the influent (0.1 M NaNO<sub>2</sub>) and the effluent. The collected effluent was titrated against a standard NaOH solution.

#### RESULTS AND DISCUSSION

Adsorption behaviour of different ions on the polyaniline stannic silicomolybdate

- a) Initial rate of exchange is proportional to concentration of  $^{137}$ Cs more than  $2x10^{-2}$  M, and becomes at constant above this concentration.
- b) Diffusion control of solution depend on the particle size.
- c) Distribution coefficients ( $K_d$  for polyaniline stannic silicomolybdate increase by increasing the metal concentration according equation (1). Show in. Figures 3-6,. Where  $A_i$  and  $A_f$  are initial and final concentrations in mol/g
- d): Calculation of the diffusion cations of the diffusion cations usually requires the knowledge of its distribution coefficient  $(k_d)$  which was calculated from the measured concentrations in the aqueous phase according to eqution (2).:
- e) Calculated by using Freundlich isotherm parameters. On the effect of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu concentration on their uptake was investigated in the concentration range. 2x10<sup>-5</sup>- 2x10<sup>-2</sup> M.

#### Freundlich Adsorption Isotherm

This is commonly used to describe the adsorption characteristics for the heterogeneous surface

(26). These data often fit the empirical equation proposed by Freundlich. Figure 4-6: The Freundlich isotherm studies were conducted in order to determine the maximum adsorption capacity of polyaniline stannic silicomolybdate, towards <sup>137</sup>Cs, <sup>90</sup>Sr & <sup>152+154</sup>Eu. The Freundlich isotherm applies to adsorption on completely homogenous surfaces with negligible interaction between adsorbed molecules. This theory considers that all surface sites have the same adsorption energy(27). The relation between the amount of adsorbate-adsorbed by the adsorbent can be expressed by the linearized Freundlich adsorption isotherm as:

$$\mathbf{Q}_{e} = \mathbf{K}_{f} \mathbf{C}_{e}^{1/n} \tag{2}$$

Where  $K_f$  = Freundlich isotherm constant (mg/g) n = adsorption intensity;  $C_e$  = the equilibrium concentration of adsorbate (mg/L)  $Q_e$  = the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g). Linearizing equation (2), we have:

$$\log Q_e = \log K_f + \frac{1}{n} \log C_e \tag{3}$$

The constant  $K_f$  is an approximate indicator of adsorption capacity, while 1/n is a function of the strength of adsorption in the adsorption process(18). If n = 1 then the partition between the two phases are independent of the concentration. If value of 1/n is below one it indicates a normal adsorption. On the other hand, 1/n being above one indicates cooperative adsorption(19-29). The function has an asymptotic maximum as pressure increases without bound. As the temperature increases, the constants kand n Eq(3), the change to reflect the empirical observation that the quantity adsorbed rises more slowly and higher pressures are required to saturate the surface. However,  $K_f$  and n are parameters characteristic of the sorbent-sorbate system, which must be determined by data fitting and whereas linear regression is generally used to determine the parameters of kinetic and isotherm models(20). Specifically, the linear least-squares method and the linearly transformed equations have been widely applied to correlate sorption data where 1/n is a heterogeneity parameter, the smaller 1/n, the greater the expected heterogeneity. This expression reduces to a linear adsorption isotherm when 1/n = 1. If n lies

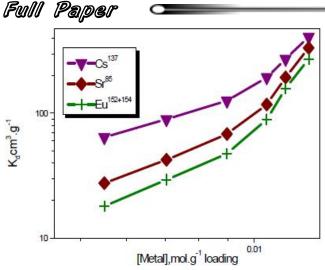


Figure 3: Change of the distribution coefficient depending on the metal concentration loading in chloride medium

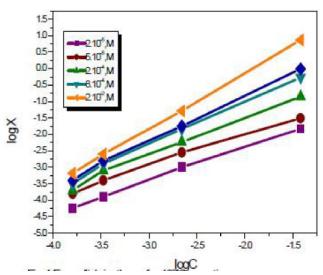


Figure 5: Freundlich isotherm for 85Sr sorption on polyaniline stannic silicomolybdate

between one and ten, this indicates a favorable sorption process(21-24). From the data in TABLES (1-3), that value of 1/n = 0.82 while n=0.75, 0.84,0.89 respectively. Indicating that the sorption of  $^{137}$ Cs,  $^{85}$ Sr and  $^{152+154}$ Eu on to polyaniline stannic silicomolybdate) are favorable and the  $R_1$ ,  $R_2$  and  $R_3$ =0.966, 0.960and 0.9598 respectively. The Freundlich parameters were used in the calculation of the site distribution function for an exchange of  $^{137}$ Cs,  $^{85}$ Sr and  $^{152+154}$ Eu. The site distribution function is given by Sposito(25) as:

$$\frac{m_{(q)}}{m_{max}} = \frac{2\cos(\pi n)\exp[n(q_m - q)] + 2\exp[n(q_m - q)]}{1 + 2\cos(\pi n)\exp[2n(q_m - q)]}$$
(4)

Where  $m_q/m_{max}$  is the ratio of the number of sites of

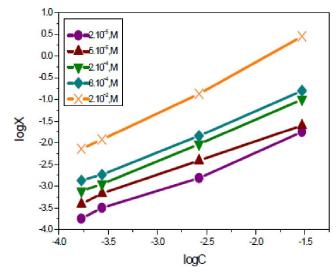


Figure 4: Freundlich isotherm for 137Cs sorption on polyaniline stannic silicomolybdate

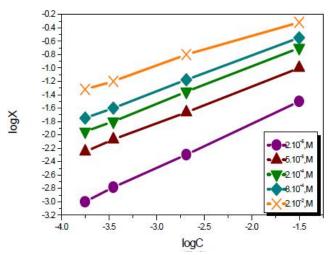


Figure 6 : Freundlich isotherm for 152+154Eu sorption on polyaniline stannic silicomolybdate

class q to the value of  $m_{(q)}$  at its maximum. The parameter q representing the class of adsorption sites is defined as:

$$q = \ln(K_{\Delta}/K_{R}) \tag{5}$$

Where  $K_A$  and  $K_B$  are the affinity parameters for the competing species A vs. B; qm is the value of q when  $m_q = m_{max}$  and is calculated by Eq(6):

$$q_{m} = \frac{1}{n} \ln(KC_{B}^{n}/M) \tag{6}$$

Where M is the total number of adsorption sites and  $C_B$  is the concentration of the species B. By assuming that the M value is equal to the maximum sorption capacity  $^{137}\text{Cs}>^{85}\text{Sr}>^{152+154}\text{Eu}$ .

#### Dubinin-Radushkevich isotherm model

TABLE 1: Freundlish and D-R parameters of CsCl on Polyaniline stannic silicomolybdate

Freundlish				D-R			
CsCl,M	X10 <sup>2</sup> k	n	R	x10 <sup>3</sup> X <sub>m</sub> molg <sup>-1</sup>	$X 10^2 k$	EkJ.mol <sup>-1</sup>	R
2.10 <sup>-5</sup>	4.05	0.69	0.98	4.86	-10.50	18.1	0.983
$2.10^{-4}$	6.12	0.51	0.97	7.35	-9.20	18.5	0.991
$2.10^{-3}$	5.78	0.62	0.95	6.84	-8.52	18.7	0.975
$2.10^{-2}$	6.25	0.75	0.94	7.50	-6.97	19.1	0.999
$2.10^{-2}$	6.75	0.77	0.98	8.1	-6.16	19.2	0.988

TABLE 2: Freundlish and D-R parameters of SrCl, on Polyaniline stannic silicomolybdate

Freundlish				D-R			
SrCl <sub>2</sub> ,M	X10 <sup>2</sup> k	n	R	x10 <sup>3</sup> X <sub>m</sub> molg <sup>-11</sup>	$X10^2 k$	EkJ.mol <sup>-1</sup>	R
2.10 <sup>-5</sup>	5.06	0.79	0.922	6.87	-10.98	18.79	0.998
$2.10^{-4}$	7.65	0.81	0.894	9.18	-9.00	18.99	0.987
$2.10^{-3}$	7.12	0.85	0.989	8.55	-8.99	19.30	0.999
$2.10^{-2}$	7.81	0.87	0.988	9.37	-8.55	19.50	0.997
2.10-2	8.44	0.9	0.899	10.13	-7.99	19.80	0.979

TABLE 3: Freundlish and D-R parameters of EuCl<sub>3</sub> on Polyaniline stannic silicomolybdate

Freundlish				D-R			
EuCl <sub>3</sub> M	X10 <sup>2</sup> k	n	R	x10 <sup>3</sup> Xnmolg <sup>-1</sup>	$X10^2k$	EkJ.mol <sup>-1</sup>	R
1	6.32	0.86	0.929	8.59	-11.20	18.91	0.988
$5.10^{-1}$	9.57	0.89	0.899	11.48	-9.5	18.95	0.999
$5.10^{-2}$	8.91	0.90	0.994	10.69	-8.75	19.50	0.989
$5.10^{-3}$	9.76	0.91	0.989	11.71	-8.55	19.78	0.997
$5.10^{-4}$	10.55	0.93	0.988	11.66	-8.02	19.80	0.999

Dubinin-Radushkevich isotherm is generally applied to express the adsorption mechanism with a Gaussian energy distribution onto a heterogeneous surface(19). Eq(7,8). The model has often successfully fitted high solute activities and the intermediate range of concentrations data well in Figure,s (7-9).

$$\mathbf{q}_{a} = (\mathbf{q}_{c}) \exp(-\mathbf{K}_{ad} \mathbf{\epsilon}^{2}) \tag{7}$$

$$\ln q_e = \ln(q_s) - (K_{ad} \epsilon^2)$$
 (8)

Where  $q_e$ ,  $q_s$ ,  $K_{ad}$ , are  $q_e$  = amount of adsorbate in the adsorbent at equilibrium (mg/g);  $q_s$  = theoretical isotherm saturation capacity (mg/g);  $K_{ad} = Dubinin-$ Radushkevich isotherm constant (mol<sup>2</sup>/kJ). The approach was usually applied to distinguish the physical and chemical adsorption of metal ions with its mean free energy,eq(9). E per molecule of adsorbate (for removing a molecule from its location in the sorption space to the infinity) can be computed by the relationship:

$$\mathbf{E} = \left[ \frac{1}{\sqrt{2\mathbf{B}_{\mathrm{DR}}}} \right] \tag{9}$$

Where  $B_{DR}$  is denoted as the isotherm constant. Mean while, the parameter can be calculated as Eq (10):

$$\varepsilon = RT \ln \left[ 1 + \frac{1}{C_c} \right] \tag{10}$$

From the linear plot of, was determined to 0.42,  $0.78 \& 0.94.6 \text{ for } ^{137}\text{Cs} < ^{85}\text{Sr} < ^{152+154}\text{Eu respectively.}$ 

### Separation of radioactive isotopes which sorbed on impregnated solid support

Hexacyanoferrate (II) Impregnated Resins Polyaniline stannic silicomolybdate (PASSM) high surface area, macroporous, resins which have been physically impregnated and it is dissolved in water. Hexacyanoferrate (II) (Sigma-Aldrich), Figure (10) shown selectivity to <sup>137</sup>Cs. The ability of the resins to recover 137Cs /HCl from solution has been investigated.. The overall efficiency of the resin was

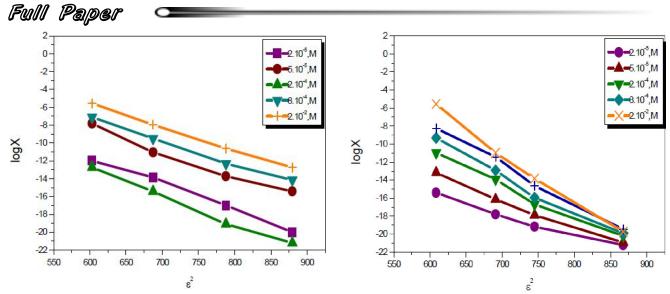


Figure 7: D\_R isotherm for for 137Cs sorption on polyaniline stannic silicomolybdate

Figure 8: D\_R isotherm for for 85Sr sorption on polyaniline stannic silicomolybdate

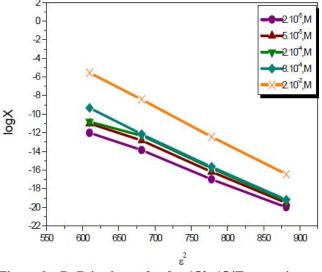


Figure 9: D\_R isotherm for for 152+154Eu sorption on polyaniline stannic silicomolybdate

found to be dependent on pH (1.5-2). It is also related to the level of impregnation of the resins. The recovery by the resin with Solid/liquid as column ion exchange using the same solvent was done. The capacity of the resin was estimated by determination of the maximum <sup>137</sup>Cs, <sup>85Sr</sup> and <sup>(152+154)</sup>Eu loading of the resin, Figure(10). The adsorption of <sup>137</sup>Cs with solvent impregnated hexacyanoferrate (II), progresses via Longmuir adsorption mechanism and high maximum adsorption of (0.988mmol/g) is obtained at pH~ 3.5. The resin impregnated in hexacyanoferrate (II), were prepared using extractant adsorption properties for <sup>137</sup>Cs higher capacity than stannic resin and due to less leakage of extrac-

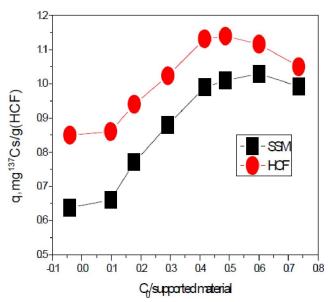


Figure 10 : Comparison of adsorption capacity between PASSM & HCF

tant. 45% higher impregnated capacity for hexacyanoferrate (II),. the conventional resin and a maximum adsorption capacity of 2.5 meq/g for <sup>137</sup>Cs was obtained 3 times.

#### Elution if radioactive isoropes

For the elution of <sup>137</sup>Cs from solid phase, the same method was applied using a solution of 0.03 M EDTA +0.1 NaNO<sub>3</sub> at pH~ 6.53 was passed through the column at the same flow rate (2ml/min)as with the batch experiments, the effluent analysis showed that <sup>137</sup>Cs, where <sup>137</sup>Cs >85Sr and <sup>(152+154)</sup>Eu is eluted respectively. Figure,s 11 2.10-2M of nitric acid

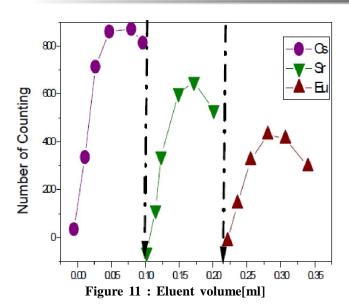


TABLE 4: Separation feasibility of fission products from each other by polyaniline stannic silicomolybdate from hydrochloric acid medium

HCl	Cs <sup>+</sup> /Sr <sup>2+</sup>	<sup>2</sup> +Sr/ <sup>3+</sup> Eu	+Cs/3+Eu
	1.33	1.52	1.92
$\mathbf{M}^{+}$	$10^{-2}$	$4x10^{-4}$	$2x0^{-5}$

used as eluent for removal and separation process. The preconcentration and removal of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu radioactive waste was done by recovery percent more than 94.6% for <sup>137</sup>Cs 78% for <sup>85</sup>Sr and 42% for <sup>152+154</sup>Eu.

#### **Separation feasibility**

Study the distribution behaviour of radioactive waste on polyaniline stannic silicomolybdate from HCl medium, separation of <sup>137</sup>Cs >85Sr and (152+154)</sup>Eu from each other depending on the separation factor evaluated in TABLE 4.

#### **CONCLUSION**

In the present study, Adsorption of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>(152+154)</sup> Eu on polyaniline Stannic Silicomolybdate adsorbent in aqueous chloride medium has been prepared with a good ion exchange capacity (2.5meq/g for polyaniline stannic silicomolybdate. The adsorption increase in the presence of NaCI. Chemical analysis chromatography study the Dubinin–Radushkevich isotherm model and Freundlich adsorption Isotherm shows that The Freundlich parameters were calculated and using in

the calculation of the site distribution function for an adsorption of <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu. From the calculated R for Freundlich and Radushkevich, it is found that R,s for <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>152+154</sup>Eu is higher for D-R isotherm than Freundlich adsorption Isotherm: model. The radioactive isotopes <sup>137</sup>Cs, <sup>85</sup>Sr and <sup>(152+154)</sup>Eu were separated selectively. Adsorption studies showed the material to be selective for <sup>137</sup>Cs ion, which is a major polluting radionuclide in many nuclear industry effluents. The polyaniline stannic silicomolybdate exchanger effectively separates <sup>137</sup>Cs ions from other fission and activation products (Sr, Cs, and Eu). A special feature is that it has potential for decontaminating acidic waste solutions, which cause severe environmental problems.

#### REFERENCES

- [1] A.A.Khan, M.M.Alam, (55), 277-290 (2003).
- [2] M.Qureshi, J.P.Gupta, V.Sharma, (21), 102-106 (1974).
- [3] S.A.I.Khan, A.A.Khan, Talanta, (71), 841-847 (2007).
- [4] H.M.F.Freundlich; "Leipzig, (57), 385-470 (1906).
- [5] B.Al-Duri; In: G.McKay, Edition, CRC Press, 133 (1996).
- [6] D.O.Campbell, T.A.Dillow, J.IAEA-TEC DOC-675.IAEA.Vienna., 85-106 (1992).
- [7] Richard Schiffman, "Two years on, America hasn't learned lessons of Fukushima nuclear disaster", (2013).
- [8] Martin Fackler; "Report Finds Japan Underestimated Tsunami Danger, (2011).
- [9] Operational monitoring good practice guide "The selection of alarm levels for personnel exit monitors" National Physical Laboratory, Teddington UK, Dec, (2009).
- [10] T. Yassine J. Radioanalytical Chem., (13), (2005).
- [11] Syed Ashfaq Nabi, Rani Bushra, Mohammad Shahadat, Journal: Toxicological & Environmental Chemistry, (1), 1-14 (2012).
- [12] Z.Hubicki, P.Podkościelny, E.Robens; Journal: Chemosphere, (56), 91-106 (2004).
- [13] A.P.Gupta, G.L. Verma, Saiqa Ikram; Journal: Reactive & Functional Polymers, (43), 33-41 (2000).
- [14] Qin Li, Jianping Zhai, Wenyi Zhang, Mingmei Wang, Jun Zhou; Journal of Hazardous Materials, (141), 163-167 (2007).

- [15] F.A.Miller, C.H.Wilkins; Journal: Analytical Chemistry, (24), 1253-1294 (1952).
- [16] S.A.Nabi, R.Bushra, Z.A.Al-Othman, Mu.Naushad; Journal: Separation Science and Technology, (46), 847-857 (2011).
- [17] M.I.Tempkin, V.Pyzhev; Acta Phys.Chim.USSR, (12), 327–356 (1940); (A) Derylo-Marczewska, M.Jaroniec<sup>1</sup>, J.Os'cik, A.W.Marczewski; Correlations among parameters of Dubinin-Radushkevich and Langmuir-Freundlich isotherms for.
- [18] M.M.Dubinin; Chem.Rev., 60, 235–266 (1960).
- [19] Syed Ashfaq Nabi, Sajad Ahmad Ganai, Amjad Mumtaz Khan; Journal of Inorganic and Organometallic Polymers and Materials., (21), 25-35 (2011).

- [20] A.Dabrowski, Adsorption—from theory to practice, Adv.Colloid Interface Sci., 93, 135–224 (2001).
- [21] M.M.Dubinin; Chem.Rev., 60, 235–266 (1960).
- [22] J.P.Hobson; J.Phys.Chem., 73, 2720–2727 (1969).
- [23] K.Y.Foo; B.H.Hameed; Chemical Engineering Journal, **156**, 2–10 (**2010**).
- [24] S.Goldberg; Chemical Processes in Soils, SSSA Book Series, 8, (2005).
- [25] Freundlich HMF.Über die.Z Phys Chem., 57(A), 385–470 (1906).
- [26] B. Volesky; Hydrometallurgy, **59**, 203–216 (2001).
- [27] A.K.Bhattacharya, S.N.Mandal, S.K.Das; Journal Chem.Eng., 123, 43–51 (2006).
- [28] P.C.Mishra, R.K.Patel; J.Hazard.Mater., 168, 319–325 (2009).